

Effective tight-binding model for renormalized band structure of Sr_2RuO_4

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We derive an effective quasiparticle tight-binding model which is able to describe with high accuracy the low-energy electronic structure of Sr_2RuO_4 obtained by means of low temperature angle resolved photoemission spectroscopy. Such approach is applied to determine the momentum and orbital dependent effective masses and velocities of the electron quasiparticles close to the Fermi level. We demonstrate that the model can provide, among the various computable physical quantities, a very good agreement with the specific heat coefficient and the plasma frequency. Its use is underlined as a realistic input in the analysis of the possible electronic mechanisms related to the superconducting state of Sr_2RuO_4 .

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Since its discovery, the nature of the superconducting state of Sr_2RuO_4 remains in the focus of the solid state research [1–4]. An accurate description of the low energy electronic structure is a fundamental step for understanding the collective properties of complex materials. This is also the case for the superconducting phase of Sr_2RuO_4 . There are generally two ways to get access at the electronic structure of a given material. On one side, *ab initio* density functional theory (DFT) can provide quasiparticle spectrum at all energies, although it is known to be not suitable for properly accounting the effects of electron correlations. To this end, DFT calculations are often taken as a platform for a more elaborate treatment of correlation effects as, for instance, in DFT+DMFT (dynamical mean-field theory) approaches, or other many-body theories. Such methods, in the attempt to build up an accurate quantitative description of correlated materials, usually includes the Coulomb interaction within tight-binding (TB) models based on a localized Wannier basis from the DFT states. The complexity in dealing with the high and low energy sector of correlated materials on equal footing leads to deviations between the theoretical predictions and the experimental observations. These can manifest themselves, for instance, in the difficulty to capture the observed band renormalization, to quantitatively reproduce the relative band positions [5], *etc.*

On the other hand, there are different experimental methods to probe directly and indirectly the electronic

structure. For instance, the thermodynamical properties can provide average information on the physical quantities at the Fermi level (FL) such as density of states. Otherwise, by means of de Haas–van Alphen or Shubnikov–de Haas measurements via the analysis of the resonance frequencies of the cyclotron motion it is possible to map the Fermi surface and to extract the effective masses at the FL, assuming that suitable conditions for the applied magnetic field and the degree of purity of the samples are given. For the Compton scattering probe, which recently gained popularity with layered superconductors, one has to face the reconstruction of a 2D electron density from a set of experimentally measured Compton profiles [6–11]. In this framework, in terms of band mapping [12], angle-resolved photoelectron spectroscopy (ARPES) appears to be the most direct momentum and energy resolving technique for determination of the electronic structure.

Concerning the Sr_2RuO_4 , though for the first ARPES measurements it was not easy to disentangle the contributions of the surface states from the bulk ones [13], the improvement of the experimental analysis allowed to get a general agreement between photoemission and bulk probes [6, 15]. Interestingly, the recent observation of an anomalous splitting of the β surface bands renewed interest in the study of Sr_2RuO_4 electronic structure [14]. While various reports on integrated quantities (like average Fermi velocities or effective masses) characterizing the band structure of Sr_2RuO_4 are available in the literature, a detailed quantitative description of the low energy electronic structure of Sr_2RuO_4 as measured by ARPES is still missing. In this paper, starting from low temperature high resolution ARPES observations, we aim at providing an effective TB model to quantitatively describe the dispersion of the renormalized low energy quasiparticles of

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Sr_2RuO_4 , following an approach that is similar to what has already been done for the layered dichalcogenides $2H\text{-TaSe}_2$ and $2H\text{-NbSe}_2$ [16, 17].

Effective TB models, i.e. a representation of the electronic structure within a certain energy region close to the FL in terms of atomic-like orbitals, is a powerful method often used to analyze the essential mechanisms governing the physical behavior of complex materials. Moreover, one of the basic advantages of a TB model is that it allows the band structure to be computed on very fine meshes in the Brillouin zone at low computational cost, which, furthermore, greatly facilitates calculation of transport, superconducting and other properties determined by peculiarities of the Fermi surface and the dispersion of low energy electronic bands [18].

TB models with the corresponding sets of parameters as derived from the first-principles calculations of Sr_2RuO_4 have been reported earlier [18–24], and used to calculate the magnetic response [21, 25], the Hall coefficient [23] and the photoemission spectra [20]. Unlike the previous examples, where unrenormalized band structure was captured, TB models were also successfully applied to parameterize the dynamics of quasiparticles, as in the case of graphene [26], for the reconstructed diamond surface $\text{C}(111)2\times 1$ [27] or in iron arsenides [28]. Here we combine our experimental data with a quasiparticle tight-binding approach to produce an accurate description of quasiparticle dispersion in single layer ruthenate Sr_2RuO_4 in the vicinity of the FL.

High-quality Sr_2RuO_4 single crystals used in this work have been grown by the flux-feeding floating-zone technique with Ru self-flux [30, 31]. The composition and structure of the samples have been characterized by X-ray and electron backscatter diffraction. All the diffraction peaks had the expected (001) Bragg reflections of the Sr_2RuO_4 phase, confirming the absence of any spurious phase. The purity of the crystals is supported by a.c. susceptibility and resistivity measurements demonstrating a narrow superconducting transition with $T_c=1.34\text{ K}$, which is a signature of a low impurity concentration [32]. Photoemission data were collected at the BESSY 1³ ARPES station equipped with a SCIENTA R4000 analyzer and a Janis ³He cryostat [33, 34]. Further details on the experimental geometry can be found elsewhere [35, 36].

Before presenting the modeling of the Sr_2RuO_4 electronic structure it is worth pointing out a few aspects which have to be considered with care in the attempt of deriving a TB description of the experimental data [16, 17, 37, 38]. Indeed, electronic structure of Sr_2RuO_4 as seen in photoemission experiment can be regarded as a superposition of two sets of features, one corresponding to the bulk bands, and the other one to the surface bands [14, 39]. While the momentum disparity between the corresponding surface and bulk features is comparatively small at the FL, the difference becomes notable at higher binding energies because of the unequal

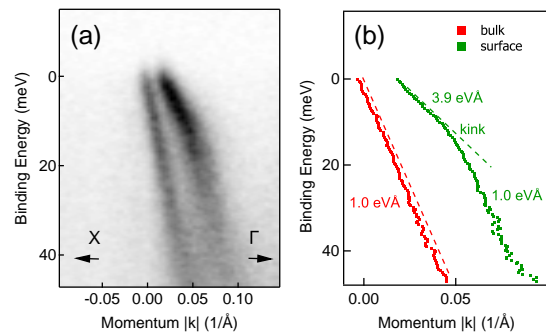


Fig. 1 Surface and bulk α bands (a) and their MDC dispersions (b). The projections of the band velocities were estimated by fitting a line to a straight segments of experimental dispersion. Since the cut is not perpendicular to the Fermi surface locus, the total in-plane velocities are actually larger.

renormalization of the surface and bulk bands [14, 35, 39]. To illustrate this issue in Fig. 1 we show a cut through the α pocket, where the surface and bulk α bands are well resolved, so that their MDC dispersions can be fit and traced down to about 50 meV in binding energy. We find that the velocity of the bulk band projected on the cut direction is about $1\text{ eV}\cdot\text{\AA}$, and does not vary much within the first 50 meV below the FL. However, for the surface band, contrary to the expectations expressed in Ref. 39, we find an abrupt change in the band velocity located at about 17 meV binding energy.

Such a kink in the band dispersion typically signals the occurrence of a coupling between electrons and bosonic modes, which at these energies are typically ascribed to phonons [40–43]. Considering the evidence for a strong electron phonon coupling in Sr_2RuO_4 , which is based on the neutron data by Braden *et al.* [44, 45] and theoretical calculations [46, 47], it is interesting to have a closer look at this issue. One may notice that up to 4 THz ($\sim 16.5\text{ meV}$) there are only acoustic phonon branches, and in the range 4–5 THz weakly dispersing optical phonons of various symmetries are present, which seem to be a good candidate to cause the observed kink in the band dispersion. Such a variation in the electron–phonon coupling for the bulk and surface bands may seem surprising at first. However, the lower symmetry of the local ionic environment is likely to account for the enhanced electron–phonon coupling at the metal surface [48, 49]. This dichotomy also helps to clarify the difference between the Ingle *et al.* [39] and Iwasawa *et al.* [50] reports, who showed a practically flat dispersion for the α band, on one side, and the kink reported by Aiura *et al.* [51] and Kim *et al.* [52] on the other side. In view of the current data we believe the latter two experiments must have been performed under conditions of a dominating surface component.

There are two outcomes from this observation. The first one, mainly pertaining to the current study, is that when constructing any TB fit intended to describe the bulk

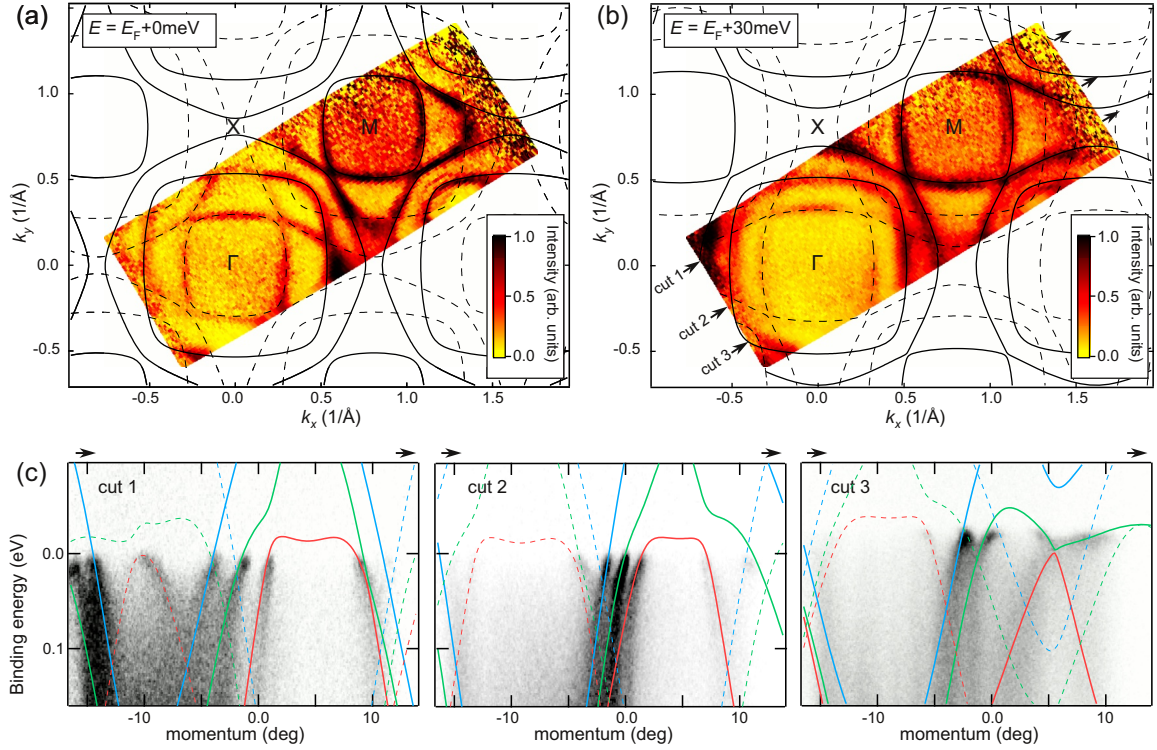


Fig. 2 (a) Experimental Fermi surface of Sr_2RuO_4 with superimposed TB contours. (b) Intensity distribution similar to the Fermi surface map shown in (a) but taken 30 meV below the FL. (c) Comparison of experimental intensity distribution for several energy-momentum cuts with fitting quasiparticle dispersion. Cuts position in momentum space is marked by small arrows in panel (b).

band structure of Sr_2RuO_4 , one always has to pick the band with a higher Fermi velocity from the two close bulk and surface features. A higher Fermi velocity for the bulk counterparts as compared to surface ones has also been observed in other layered superconductor $\text{YBaCu}_2\text{O}_{7-\delta}$ [53], which brings about the second outcome, apparently affecting band renormalization studies performed with ARPES in general. The point is that the momentum splitting between the surface and bulk bands might be negligibly small, hence treating an unresolved composite feature as a single one may lead to an underestimated Fermi velocity (overestimated renormalization) as contrasted to the true bulk values.

Let us consider the effective TB model. In comparison to many other layered superconductors Sr_2RuO_4 is known to have a relatively weak k_z dispersion [54–58], which is still further reduced by the spin-orbit interaction [59]. Therefore, in choosing an appropriate TB model for Sr_2RuO_4 we neglect the k_z dispersion and follow the basic formulation already proposed by Ng et al. as well as by other authors [2, 60–62]. In this framework, the TB Hamiltonian can be expressed as follows:

$$H = \sum_{\vec{k}, s} \psi_s^\dagger(\vec{k}) \hat{A}(\vec{k}) \psi_s(\vec{k}) + \text{h.c.}, \quad (1)$$

where $\psi_s(\vec{k}) = [d_s^{yz}(\vec{k}), d_s^{xz}(\vec{k}), d_s^{xy}(\vec{k})]^T$ indicates the ba-

sis with a three-component spinor and the matrix $\hat{A}(\vec{k}, s)$ is given by

$$\hat{A}(\vec{k}) = \begin{pmatrix} \epsilon_k^{yz} - \tilde{\mu} & \epsilon_k^{\text{off}} + i\lambda & -\lambda \\ \epsilon_k^{\text{off}} - i\lambda & \epsilon_k^{xz} - \tilde{\mu} & i\lambda \\ -\lambda & -i\lambda & \epsilon_k^{xy} - \tilde{\mu} \end{pmatrix}, \text{ and} \quad (2)$$

$$\begin{aligned} \epsilon_k^{yz} &= -2\tilde{t}_2 \cos(k_x) - 2\tilde{t}_1 \cos(k_y); \\ \epsilon_k^{xz} &= -2\tilde{t}_1 \cos(k_x) - 2\tilde{t}_2 \cos(k_y); \\ \epsilon_k^{xy} &= -2\tilde{t}_3 (\cos(k_x) + \cos(k_y)) - 4\tilde{t}_4 \cos(k_x) \cos(k_y) - \\ &\quad 2\tilde{t}_5 (\cos(2k_x) + \cos(2k_y)); \\ \epsilon_k^{\text{off}} &= -4\tilde{t}_6 \sin(k_x) \sin(k_y). \end{aligned}$$

In Fig. 2 we fit the model parameters in order to optimally reproduce the experimental data in an energy window close to the FL. Panel (a) shows the TB-model Fermi surface contours superimposed over the experimental data. The effective electronic parameters which provide the best description for the dispersion of the low energy quasiparticles in Sr_2RuO_4 can be summarized in the following table:

$\tilde{\lambda}$	\tilde{t}_1	\tilde{t}_2	\tilde{t}_3	\tilde{t}_4	\tilde{t}_5	\tilde{t}_6	$\tilde{\mu}$
0.032	0.145	0.016	0.081	0.039	0.005	0.000	0.122

Having at hand simple equations describing the band dispersion it is easy to calculate the density of states

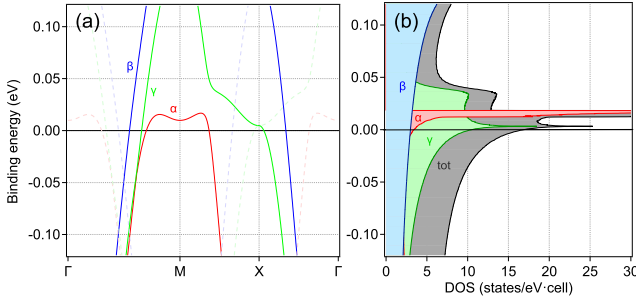


Fig. 3 Dispersion of the quasiparticle TB bands (a) and the derived quasiparticle density of states (b) in the vicinity of the FL.

(DOS) and make an estimate for the electron count and electronic specific heat [63]. To calculate DOS we used tetrahedron method of Lehmann and Taut [64]. The results are shown in Fig. 3. The estimated density of states at the FL for the three low energy bands are as follows: $g_\alpha \approx 3.4$, $g_\beta \approx 3.0$ and $g_\gamma \approx 10.5$ states/eV·molRu, which totals to $g \approx 16.9$ states/eV·molRu and translates to the Sommerfeld coefficient of 40 mJ/K²·molRu. The obtained number is in agreement with the experimentally measured values ranging from 29 to 45.6 mJ/K²·molRu [65–73], which indicates that the developed fit properly describes the dispersion of the bulk bands in the vicinity of the FL. The integral of the DOS up to the FL gives the electron count of 3.9 electrons per BZ, which again, within the experimental accuracy, agrees with the expected 4 electrons per BZ.

Electronic structure of materials is frequently discussed in terms of effective masses. Such a reduction to a single integral value m^* enables a comparison between various experimental and theoretical methods. In this context, it is useful to remind that besides the band mass tensor $m_{\mu,\nu} = \hbar^2 (\frac{\partial^2 \epsilon(\mathbf{k})}{\partial k_\mu \partial k_\nu})^{-1}$ different effective masses are frequently considered: (1) the band mass m_b as it can be obtained from the bare electronic dispersion, (2) the thermodynamic mass m^* , (3) the cyclotron resonance mass m_c , (4) the susceptibility mass m_{suscept}^* , (5) the plasma frequency mass m_p [15, 74]. Here, the quasiparticle densities of states can be easily recalculated into thermodynamic masses ($m^*/m_e = \pi \hbar^2 g / m_e a^2$) and compared, on the band by band basis, to the thermodynamic masses reported in de Haas–van Alphen measurements as summarized in the table below.

mass type	α	β	γ	total	year
(this study)	5.4	4.8	16.7	26.9	
Cyclotron thermodynamic [15, 75]	3.3	7.0	16.0	26.3	2001
Cyclotron thermodynamic [72]	3.4	7.5	14.6	25.5	1998
Cyclotron thermodynamic [76]	3.4	6.6	12.	22.0	1998
Cyclotron thermodynamic [77]	3.2	6.6	12.0	21.8	1996
Cyclotron resonance [15]	2.1	4.3	5.8	12.2	2003
Cyclotron resonance [78]	4.3	5.8	9.7	19.8	2000

As one may notice there is a gradual increase of the total thermodynamic mass reported by the de Haas–van Alphen measurements with time, which is probably related to the improving quality of the available crystals. Our total mass ($26.9 m_e$) is closest to the most recent dHvA value of $26.3 m_e$. Similarly, we find a good correspondence for the mass of the γ band, however there seems to be a difference in the masses of α and β bands. While in the current fit these bands have practically the same mass, in the de Haas–van Alphen data their mass ratio is about two. In the two-dimensional case the density of states, and hence the effective mass, is inversely proportional to the Fermi velocity and directly proportional to the length of the Fermi surface contour. Therefore, assuming equal velocities for the α and β band would give a mass of the β band to be twice that of the α band, as the average radius of the β band is about twice as high. As clarified in ref. 15 (p. 686) this assumption was ‘actually used ... as a guiding line’ when extracting the susceptibility effective mass. Nevertheless, as can be seen from Fig. 3a, the quasiparticle Fermi velocity of the α band is systematically lower than that for the β band, thus the accurate account of the variation of the Fermi velocity, v_F , and k_F yields about the same thermodynamic masses for the α and β bands. We expect that the value extracted from the presented TB model correctly reproduces the relation between the effective mass of the α and β bands.

Besides the heat capacity, the obtained effective TB model can be used to calculate other averaged properties over the Brillouin zone, such as the plasma frequency, whose value is given by

$$\hbar\Omega_{xx} = \sqrt{\sum_{i=\alpha,\beta,\gamma} \frac{e^2}{L_a L_b L_c \epsilon_0} \left\langle f_k \frac{\partial^2 E^{(i)}}{\partial k_x \partial k_x} \right\rangle_{\text{BZ}}}, \quad (3)$$

from where we get $\hbar\Omega_{xx} = 1.3$ eV. As expected the value is about 4 times smaller than the one obtained based on the unrenormalized band structure calculation [79].

In conclusion, we have developed an effective tight-binding model that is able to capture the low energy electronic features of the quasiparticle spectra of Sr₂RuO₄ taking ARPES data as an input. Owing to different degree of renormalization in the bulk and at the surface, the bulk bands have been properly selected and analyzed for the determination of the quasiparticle model. We have extracted the momentum and orbital dependence of the Fermi velocity and of the effective masses close to the FL. As a demonstration of the use of the derived model, we have calculated the density of states and found a good agreement between the Sommerfeld coefficient calculated based on the obtained fit and the one directly measured. We believe that the developed model can be of value for a more realistic input to compute the orbital dependent magnetic properties in order to test, for example, the relevance of the ferromagnetic or antiferromagnetic fluctuations in settling the spin-triplet pairing

in the superconducting phase of Sr_2RuO_4 [2, 80, 81].

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